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CONDITIONING OF TRITIATED WASTES
PART II - SOLIDIFICATION AND ENCAPSULATION

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Work is continuing on the development of conditioning systems for low and intermediate level tritiated liquid and solid wastes which will prevent loss of tritium for at least 150 years. This portion of the program has concentrated on solidification and encapsulation of tritiated aqueous wastes, development of techniques for the measurement of tritium loss in air and water, and identification and evaluation of encapsulation materials. Solidification of tritiated aqueous wastes by water extendible polyester or cements resulted in average tritium releases of approximately $1\text{-}4\times10^{-1}$%/day with that from water extendible polyester being the lowest. The daily release rate is independent of initial tritium concentration in the waste form and can be reduced by a factor of 1000 by encapsulation of the waste within a 10 mm layer of water extendible polyester. Water extendible polyester is the preferred material for solidification and encapsulation of aqueous tritiated wastes and encapsulation of tritiated solids permitting release of only $3\times10^{-3}$% of the original activity over 150 years. It is expected that this program which was originally scheduled for three years can now be completed in two years with complete definition of the conditioning system including the outer package.

The Chemical Research Department has undertaken a program to develop low and intermediate level tritiated waste conditioning systems which will prevent loss of tritium or tritiated water from solidified liquids and encapsulated solids for at least 150 years. The program as illustrated in Appendix 1 was divided into six major



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|---|---|---|---|
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tasks: package materials identification, encapsulation materials identification, liquid waste conditioning, solid waste conditioning, technique development, and process equipment development.

In a preliminary investigation the literature was reviewed to identify candidate materials for the solidification of low level tritiated aqueous wastes and experiments with light water were performed/1/. Cement was selected as an absorbent due to its low water vapour pressure when fully hydrated, high water content, and low cost. It was concluded that Portland cement type 30 and high alumina cement would be suitable for the solidification of tritiated water. As neither material will retain all absorbed water molecules in a non-evaporable state, tritiated wastes solidified in this way cannot be considered immobilized. It was recommended that additional work on aqueous tritiated waste conditioning concentrate on solidification of the waste in a suitable medium and development of a system to prevent loss of tritiated water from the conditioned wastes.

In the absence of a suitable procedure for immobilization of tritiated wastes by solidification only, it is necessary to employ multiple barriers to reduce the tritium release rate to the environment to an acceptably low level. No specific guidelines exist but an arbitrary figure of 150 years has been selected for tritium containment. This period (~12 tritium half-lives) would reduce the activity to 0.03% of its original level. McKay/2/ considered the immobilization of tritiated wastes from fuel reprocessing and argued that the immobilized waste form need retain its integrity for only 82 years providing that the postulated constant percentage leakage of tritium not exceed $1 \times 10^{-3}$% per day. Therefore, to be acceptable, the conditioned waste must not release tritium to the surrounding medium (air or water) at a rate greater than this maximum.

Work in this portion of the program has concentrated on solidification and encapsulation of tritiated aqueous wastes, development of techniques for the measurement of tritium loss in air and water, and identification and evaluation of encapsulation materials.

In addition, in a separate project, techniques have been developed for solidifying radioactive waste oil which should also be adequate for tritiated organic liquids/3/. In parallel with the waste conditioning studies, the design and construction of a prototype radioactive waste solidification plant which will be suitable for tritiated liquid waste processing is underway in conjunction with Nuclear Materials Management Department.

## EXPERIMENTAL

### Solidification of Tritiated Aqueous Wastes

Test specimens were prepared using Portland cement type 30, high alumina cement, and water extendible polyester (WEP). This last material was included because it can accept a substantial quantity of water and cures to a hard, dry product. The water extendible polyester was originally evaluated in research on the immobilization of aqueous decontamination wastes as a result of which it has been selected for use at Darlington GS. Complete details on the preparation of the 50 mm right cylindrical specimens are presented in Appendix 2 and are summarized in Table 1.

### Encapsulation of Solidified Specimens

As part of the multiple barrier system to prevent loss of tritiated water from solidified liquids and other contaminated solids, it was postulated that completely surrounding the solid material with a liquid which would solidify would assist in preventing tritium escape.

Thermoplastic and thermosetting organic resins were considered as possible candidates. Thermoplastic resins which are solid at normal temperatures must be heated to liquify. To prevent loss of tritiated water during encapsulation, thermoplastic materials of relatively low melting points were investigated, eg, asphalt, paraffin wax, and certain polyethylenes. Asphalt and polyethylene require temperatures well above 100°C to produce free flowing liquids but it was possible to encapsulate specimens with a paraffin wax which melted at ∿70°C. Upon cooling, the wax envelope shrank slightly leaving an air gap between the encapsulant and the solid specimen. Thermoplastic resins were not investigated further for this application.

Thermosetting resins such as epoxies and polyesters are liquids at normal temperatures and solidify by the addition of a catalyst or cure agent. As a result of the curing reaction the temperature of the material generally increases to a maximum before returning to ambient. It is possible to minimize the peak exotherm temperature by the proper selection of resin, cure agent, catalyst, or filler, eg, sand, water.

Epoxy and polyester resin systems have a high degree of water resistance but in initial trials attempts to encapsulate one day old tritiated cement specimens caused bubbles to appear around the cement specimen and be trapped in the resin. The bubbles were apparently due to water on the surface of the cement being vapourized. Several epoxy and polyester systems with relatively low peak exotherm temperatures were identified which did not cause bubble formation. Along with Portland cement type 30 and water extendible polyester (WEP), they were selected as encapsulating agents (Table 2 and Appendix 3).

Three techniques were employed for evaluating the effectiveness of solidification on the immobilization of tritiated water: leaching in light water at 23°C, exposure in static air, and exposure in flowing air at 20°C, 20% relative humidity and 50 mL/min. To date only leaching has been applied also to encapsulated specimens. Details are presented in Appendices 4, 5 and 6.

## TABLE 1

### SOLIDIFIED TRITIATED AQUEOUS WASTE SPECIMENS TYPICAL VALUES

| Specimen Description | 50 mm right cylinders | | |
|---|---|---|---|
| Solidification Medium | Portland Cement Type 30* | High Alumina Cement | Water Extendible Polyester (WEP) Aropol 661-P |
| Tritiated Water Concentration µCi/mL | 50 | 50 | 50 |
| Solidification Medium mass (g) | 156 | 162 | 46 |
| Tritiated Water mass (g) | 44 | 48 | 54 |
| Water, mass fraction g water/g of product | 0.22 | 0.23 | 0.55 |
| Water, volume fraction mLwater/mL of product | 0.45 | 0.49 | 0.55 |
| Initial Tritium Concentration per Specimen (µCi) | 2200 | 2400 | 2700 |
| Cure times (days) | 1<br>28 | 1<br>28 | 1<br>- |

* To achieve proper mixing a plasticizing additive was added to the Portland cement at 2 g of plasticizer per 100 g of cement.

## TABLE 2

## ENCAPSULATED SOLIDIFIED TRITIATED AQUEOUS WASTE SPECIMENS - TYPICAL VALUES

| Specimen Description | Solidified specimens are 30 mm right cylinders surrounded by a 10 mm thick layer of encapsulating material to produce a 50 mm right cylinder | |
|---|---|---|
| Solidification Medium | Portland Cement Type 30 | Water Extendible Polyester (WEP) |
| Tritiated Water Concentration µCi/mL | 200 | 200 |
| Solidification Medium mass (g) | 39 | 15 |
| Tritiated Water mass (g) | 11 | 15 |
| Water Mass Fraction g water/g of solidified product | 0.22 | 0.50 |
| Initial Tritium Concentration per Specimen µCi | 2200 | 3000 |
| Cure times for Solidification Medium (days) | 1 | 1 |
| Encapsulation Media | Epoxies: Epon 815/506, Epon 815/Lo/Los<br>Polyesters: Aropol 661-P Water Extendible Polyester<br>Aropol C-308 filled with sand and bentonite<br>Cement: Portland type 30 | |
| Cure times for encapsulation media (days) | 7 | 7 |
| Volume of Tritiated Water/Volume of Encapsulated Product mL/mL | 0.11 | 0.15 |

## RESULTS

The percentage cumulative tritium releases from duplicate solidified tritiated specimens exposed in water for 180 days show that water extendible polyester is superior to Portland cement type 30 and high alumina cement with respect to retention of tritiated water (Figure 1, Appendix 7-1). The two cements are similar but the shape of the release curves are different. The release curves for both high alumina cement sets (one day cure and twenty-eight day cure) appear to have reached a maximum at ∿75% tritium loss. Both Portland cement sets (one day cure and twenty-eight day cure) are continuing to lose tritium after 180 days at which time releases had totalled 63% and 57% respectively. Similarly, the water extendible polyester specimens are continuing to lose tritium,reaching 24% after 180 days. The exposure trials are continuing.

The cumulative tritium releases from Portland cement specimens and water extendible polyester specimens exposed in water are greater than those from similar specimens exposed in an air stream at 20°C and 20% relative humidity flowing at 50 mL/min (Figure 2, Appendix 7-1 and 7-5). This difference was very marked with the cement specimens during the initial stages of the leaching period but after sixty days was within 12% and it is expected that the curves will continue to converge. The difference between tritium releases from water extendible polyester specimens exposed in water and flowing air is not as pronounced as that observed for cemented specimens, being less than 1% after 60 days.

Exposure of solidified tritiated liquid wastes or contaminated solids would only occur if the outer package system were breached. Depending upon storage conditions, these solids would then be exposed to either water or air. Although water immersion initially caused higher tritium releases, it would appear that within a relatively brief period exposure to flowing air would result in behaviour similar to water immersion. This observation is applicable to solidified specimens only and has yet to be confirmed for solidified specimens surrounded by an encapsulating matrix. If confirmed, further evaluation will be restricted to water immersion only.

The curves in Figure 2 for exposure of specimens in a completely static atmosphere demonstrate that the tritium content of the air within a sealed container quickly reaches a maximum; ∿0.01% of the initial tritium content in this case. Therefore, in handling of low level tritiated waste containers, the risk to the operator if the solidified product were exposed would not be great if the container were quickly resealed. The risk would be even further reduced by encapsulation of the solidified material.

The quantity of tritium found in the air just below the lid of the test container is approximately 75% of that expected from evaporation of water into a bone dry atmosphere at constant volume, ie 0.15 µCi vs 0.21 µCi (Appendix 7-4). As these experiments were designed to simulate the static air conditions inside a container and there was no mixing of the surrounding air which initially was at 26% relative humidity at 21°C, this difference can be attributed to the moisture originally present in the air.

The effect of various encapsulating materials on the release of tritium from specimens prepared with Portland cement type 30 and water extendible polyester is illustrated in Figures 3 and 4 respectively (Appendices 7-2 and 7-3). For comparison, the results obtained with solidified specimens of these materials are included. The specimens which were solidified but not encapsulated were 50 mm right cylinders. The encapsulated specimens consisted of 30 mm right cylinders surrounded by a 10 mm layer of encapsulating material.

For both substrates the encapsulating materials can be ranked in the following order of increasing effectiveness: Portland cement type 30 < Aropol C-308 < Epon 815/506 < Epon 815/Lo/Los < Water Extendible Polyester (Aropol 661-P).

The magnitude of the effect of each encapsulating material is tabulated in Table 3 as the ratio of percentage cumulative tritium release from the non-encapsulated specimen at a given time to that of the encapsulated specimen.

TABLE 3

EFFECT OF ENCAPSULATING MATERIALS ON TRITIUM RELEASES

| Encapsulating Material | Time (days) | % Tritium Release from Portland Cement Specimens | Ratio | % Tritium Release from Water Extendible Polyester Specimens | Ratio |
|---|---|---|---|---|---|
| None | 60 | 46.9 | - | 13.0 | - |
| | 150 | 60.0 | - | 22.0 | - |
| Portland Cement | 60 | 11.4 | 4.3 | 2.9 | 4.5 |
| Aropol C-308 | 150 | 2.2 | 27 | 5.0 | 4.4 |
| Epon 815/506 | 150 | 0.25 | 240 | 0.04 | 550 |
| Epon 815/ Lo/Los | 150 | 0.21 | 286 | 0.03 | 733 |
| Water Extendible Polyester | 150 | 0.05 | 1200 | 0.02 | 1100 |

The most effective encapsulating material in each case was water extendible polyester which reduced the cumulative tritium release after 150 days by a factor of ∿1000. The two epoxy systems based on Shell Epon 815 resin also retarded tritium release considerably over both substrates but were more effective over water extendible polyester.

The Ashland Aropol C-308 solvent based polyester resin was filled with sand and bentonite to reduce the peak exotherm temperature to approximately 50°C. This composite material provided some retardation of tritium release from both substrates but as shown in Figures 3 and 4, the tritium release rate is increasing rapidly and may approach that of the non-encapsulated specimens at some future date. It is believed that the fillers in the polyester provided a pathway along which tritiated water could move once the surrounding polymer was permeated.

The use of Portland cement as an encapsulating agent reduced the quantity of tritium released after 60 days by a factor of 4-5 and it is expected that this advantage will decrease with increased exposure time.

Although water extendible polyester was the most effective encapsulating material over both substrates, the shape of the curves was considerably different. The Portland cement specimens encapsulated with water extendible polyester displayed a sharp increase in the amount of tritium released after 120 days, but after 150 days this trend appears to be levelling off. The water extendible polyester specimens encapsulated with water extendible polyester have released approximately the same amount of tritium as those with the cement substrate but did not display the same sharp increase after 120 days immersion in water. This difference cannot be fully explained at this time but additional tritium release data on these specimens will be obtained to determine the long term performance of these materials.

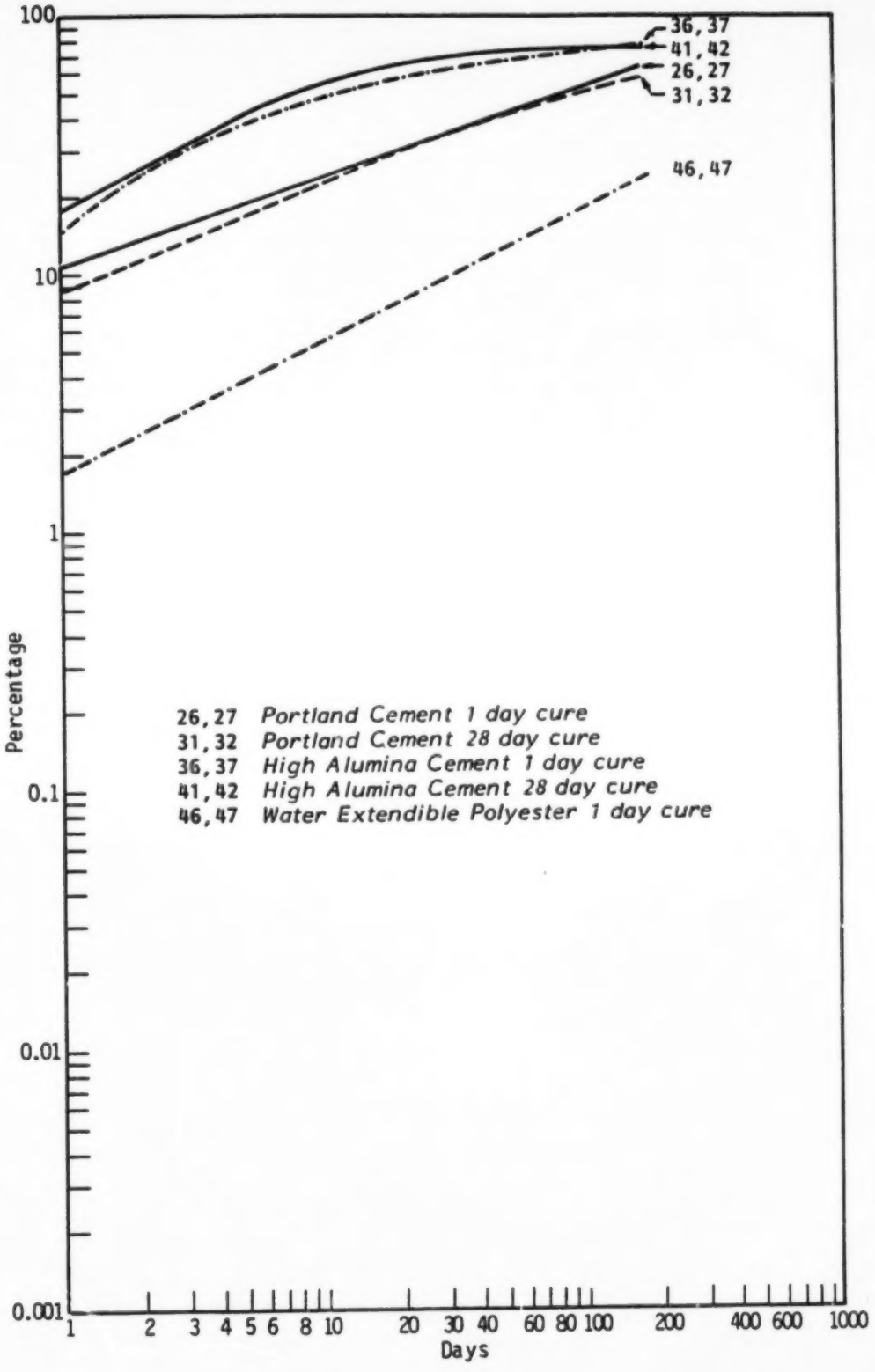


FIGURE 1

PERCENTAGE CUMULATIVE TRITIUM RELEASES IN WATER FROM SOLIDIFIED TRITIATED AQUEOUS WASTE SPECIMENS (COMPARISON OF SOLIDIFICATION MEDIA)

163949 RD

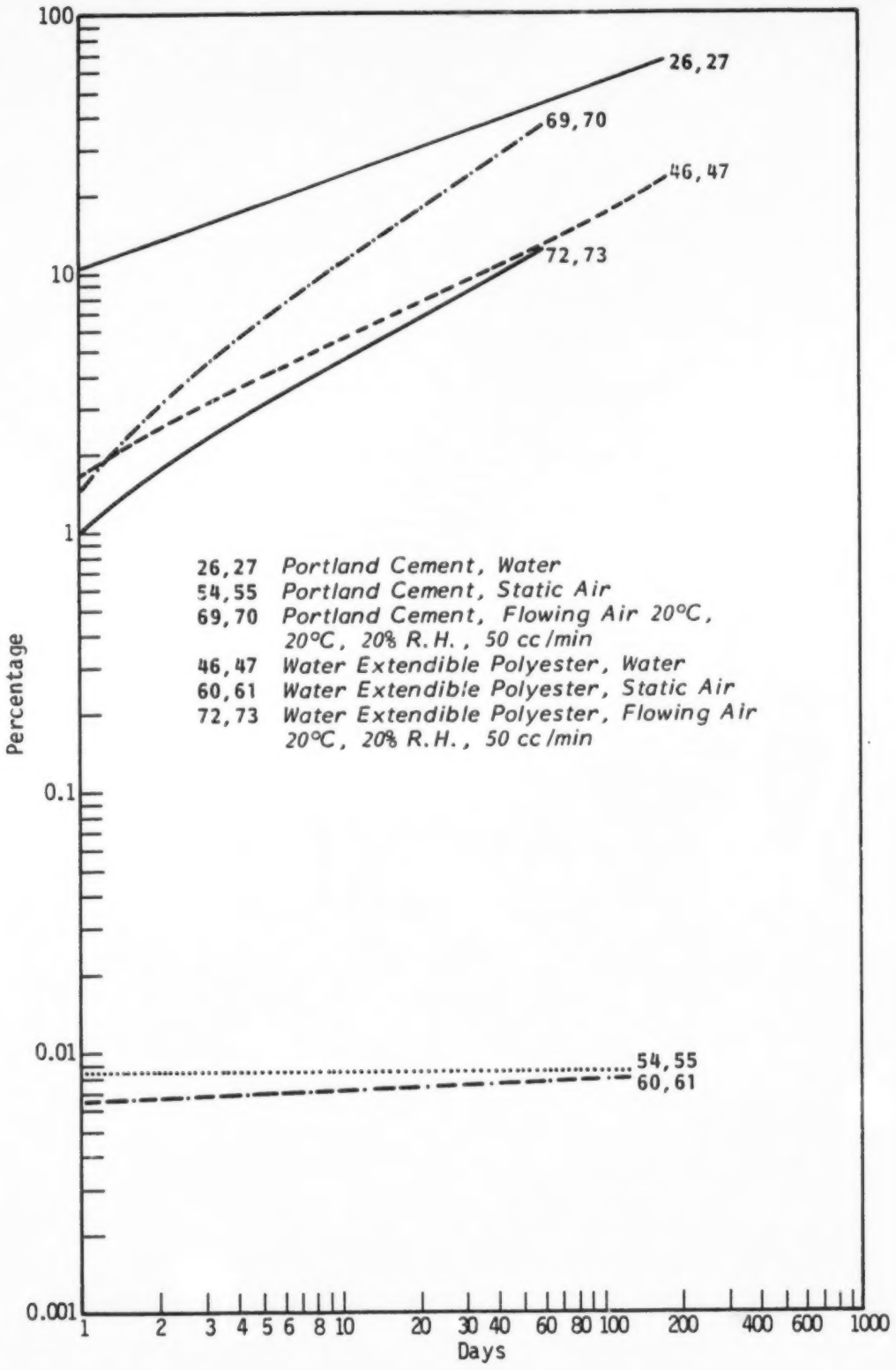


FIGURE 2

PERCENTAGE CUMULATIVE TRITIUM RELEASES IN WATER, STATIC AIR AND FLOWING AIR FROM SOLIDIFIED TRITIATED AQUEOUS WASTE SPECIMENS

163950 RD

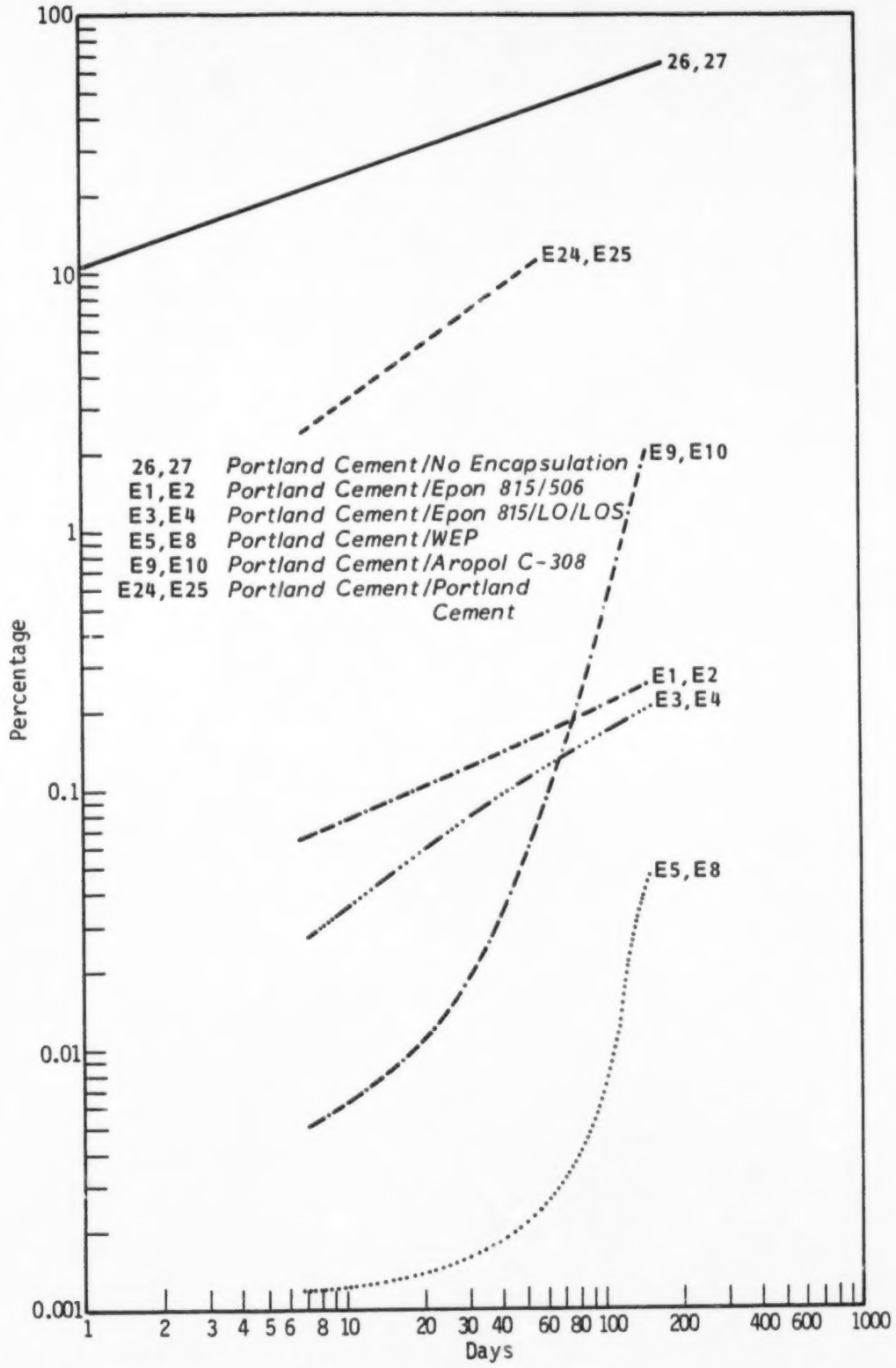


FIGURE 3

PERCENTAGE CUMULATIVE TRITIUM RELEASES IN WATER FROM SOLIDIFIED AND ENCAPSULATED TRITIATED AQUEOUS WASTE SPECIMENS (COMPARISON OF ENCAPSULATION MEDIA OVER PORTLAND CEMENT)

163951 RD

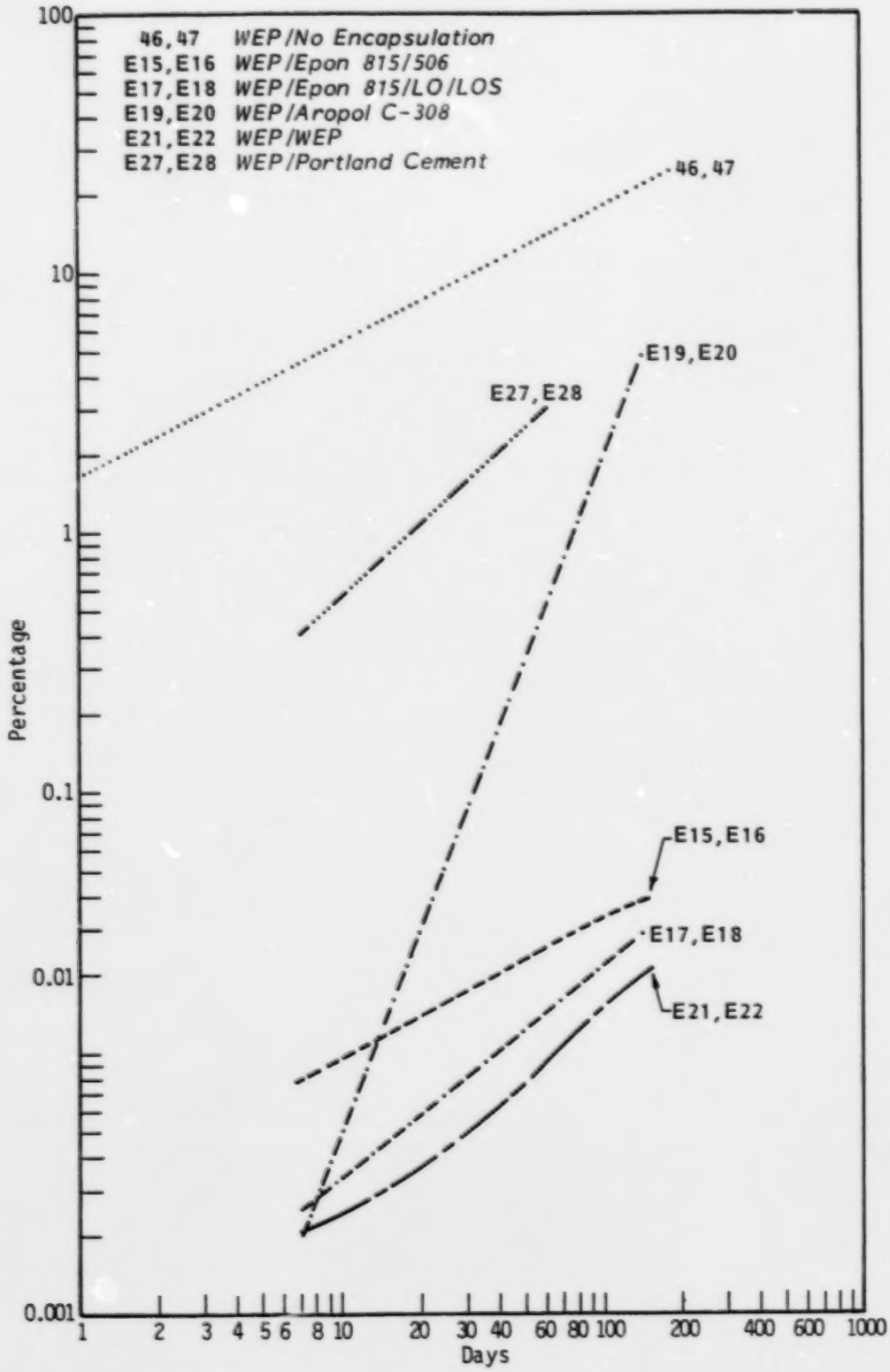


FIGURE 4

PERCENTAGE CUMULATIVE TRITIUM RELEASES IN WATER FROM SOLIDIFIED AND ENCAPSULATED TRITIATED AQUEOUS WASTE SPECIMENS (COMPARISON OF ENCAPSULATION MEDIA OVER WATER EXTENDIBLE POLYESTER [WEP])

163952 RD

## DISCUSSION

Several laboratories have investigated techniques for the immobilization of aqueous tritiated wastes with the tritium content of the waste liquids varying from 300 µCi to 300 Ci.

Pertinent data on the release of tritium during water immersion have been extracted from several sources and summarized in Table 4 along with the data obtained in this program. Where possible, similar exposure periods were selected and the tritium release data are presented as 'average daily per cent tritium released'. It is recognized that this presentation format assumes a linear relationship between time and tritium release. As was shown in Figures 3 and 4, this does not apply to some portions of this work and probably does not apply to the work at other laboratories, but it does provide a basis for comparison and is commonly used.

The average daily per cent tritium releases from those specimens which were not surrounded by some form of encapsulating material ranged from $1.3 \times 10^{-1}$ to $4.3 \times 10^{-1}$. This excellent agreement between laboratories is remarkable considering that the solidification media varied from cement to ion exchange resin in polyester and the tritium content of the aqueous waste varied from 300 µCi to 300 Ci, a factor of $10^6$. There appears to be no major advantage of one solidification media over another for the long term storage of this type of waste but water extendible polyester does provide superior initial retardation of tritium release. Tritium is unlike other radionuclides in that it does not have to be dissolved in the leachant to be mobilized as it is already part of the liquid phase and the insensitivity of the tritium release rate to such a large concentration difference suggests that it is diffusion controlled. It is necessary, therefore, to prevent by some means the egress of tritiated water or ingress of light water which may surround the waste.

The use of polymer impregnation of tritiated cement by Brookhaven National Laboratory and Mound Laboratories did not result in a consistent decrease in the tritium release rate. The specimen prepared with Portland cement only was stored in a sealed container for 462 days before being immersed in water. After 322 days in water, 81% of its original tritium content had been leached and this did not change significantly after 1000 days. The polymer impregnated tritiated cement specimen was exposed within one month of preparation and had lost 55% of its original tritium after 182 days. This increased to 87% after 1600 days. Significant improvement by a factor of 100 was obtained only when specimens of both types were sealed within polyethylene bottles before being immersed in water/5/.

The use of coatings ~1-2 mm thick was investigated at Los Alamos Scientific Laboratory but a major change in the tritium release rate was not obtained until the coating thickness was increased to 19-25 mm/6/. More recent work in France initially concentrated on absorbing the tritiated water on ion exchange resin mixing with polyester or epoxy resin and then surrounding the resulting solid with a 10 mm layer of polyester or epoxy resin/7/. The addition of this encapsulating layer reduced the tritium release rate to $\sim 10^{-4}$%/day, similar to the values found in this work with a number of substrate/encapsulating agent combinations. In a modification to their original work, tritiated water was placed inside a hollow cement cylinder whose walls were ~10 mm thick and encapsulated in a 10 mm layer of polyester or epoxy resin. The tritium release rate from the polyester encapsulated specimen was $1.8 \times 10^{-3}$%/day and from the epoxy encapsulated specimen was $1.0 \times 10^{-5}$%/day. It should be noted that by preparing specimens in this manner, the minimum pathlength from the tritium source to the surrounding media is 20 mm. This technique could not be adopted in Canada as it involves storage or disposal of a waste in the liquid state.

A similar comparison on the release of tritium from specimens exposed to air is presented in Table 5. The tritium release rate from Portland cement type 10 specimens at Los Alamos Scientific Laboratory is similar to that from Portland cement type 30 specimens and water extendible polyester specimens in this work ($2 - 6 \times 10^{-1}$%/day).

No information is yet available from this study on the behaviour of encapsulated specimens in a flowing air system but CEA (France) data show average daily tritium releases of $4.3 \times 10^{-5}$% and $2.6 \times 10^{-5}$% for epoxy and polyester encapsulating materials respectively. Unlike the results from water immersion in which the release from epoxy encapsulated specimens was less than the release from polyester encapsulated specimens by a factor of 100, the two materials behaved similarly in air exposure/7/.

On the basis of the low tritium release results obtained to date, the preferred system for conditioning of aqueous tritiated waste liquids is solidification of the waste using water extendible polyester (Aropol 661-P) surrounded by a layer of the same material prepared with light water. The use of the same material for both applications prevents damage which might occur due to differential thermal expansion of dissimilar materials. The release curve of Figure 4 does not show any significant change in slope throughout the leaching period; thus the average daily release rate of $1.4 \times 10^{-4}$%/day can be used for extrapolation over the proposed 150 year storage period. This rate compares favourably with the maximum suggested by McKay for an 82 year storage period of $1 \times 10^{-3}$%/day.

If extrapolated over 150 years with no allowance for decay, 7.7% of the tritium originally present would be released to the environment. However, the activity actually released to the environment over the 150 years is much less when allowance is made for tritium decay, according to the following equation.

$A_{150} = \lambda A_o RF$

$A_{150}$ = activity released over 150 years

$A_o$ = activity originally present in the waste form, 2860 µCi

$\lambda = 0.693/t_{1/2}$

$t_{1/2}$ = 12.3 years for tritium

RF = fractional release/year

$= 1.4 \times 10^{-4}\%/\text{day} \times 10^{-2} \times 365$

$= 5.11 \times 10^{-4}$

$$A_{150} = \left(\frac{0.693}{12.3}\right) \quad (2860) \quad (5.11 \times 10^{-4})$$

$= 8.2 \times 10^{-2}$ µCi

Therefore, the quantity of activity actually released to the environment over 150 years would be only $3 \times 10^{-3}\%$ of that originally present. As the tritium release rate appears to be independent of concentration, this would result in a total release of only 0.003 Ci for every 100 Ci originally present.

In this work, before the solidified wastes were exposed to water or air or were encapsulated, they were removed from the mold. This was necessary to evaluate the solidification and encapsulation media. It is recognized that in actual practise this would not occur as the mold would probably be retained, sealed, and act as an additional barrier to tritium release. Any encapsulating material would then be placed around the inner package. The effect of inclusion of a mold on the tritium release rate has not been investigated but may be estimated on the basis of the Mound data presented in Table 4. Retention of the specimens within a polyethylene bottle through the exposure period reduced the average daily per cent tritium release by a factor of 100. If this were applied to the already low rate of $1.4 \times 10^{-4}\%$ per day for the WEP/WEP system, the release rate could be as low as $10^{-6}\%$ per day.

In the conditioning of solid wastes such as damaged valves, etc, from the tritium recovery facility, no mold would be employed. It is proposed that the solid part would be suspended in a basket within a container and all free space filled with water extendible polyester completely surrounding the item. The effectiveness of this encapsulation procedure has been demonstrated here as encapsulation of the solidified inner specimen simulates encapsulation of solid wastes.

There is some deviation from the actual in that the tritium is dispersed evenly throughout the inner specimen but would be restricted to the surface of a damaged part.

The use of water extendible polyester as a solidification and encapsulation material for aqueous tritiated wastes makes it possible to use a single unified approach to all Ontario Hydro low and intermediate level aqueous radioactive wastes. Water extendible polyester is proposed for use at Darlington NGS for solidification of all low and intermediate level aqueous wastes which cannot be treated by ion exchange resin or dilution and dispersion and studies are underway on the use of this material for encapsulation of spent ion exchange resins for disposal.

The prototype radioactive waste solidification plant which is under construction is designed around the use of water extendible polyester resin. It is compact, sealed, and can be easily modified to permit enclosure of the appropriate components within a glove box to prevent exposure of the operator to tritiated water vapour during processing.

The research program (Appendix 1), which was originally expected to last three years, included a significant component for studying the durability of package system materials under conditions expected in storage or disposal. This would permit selection of outer package materials and assist Nuclear Materials Management Department in the design of a suitable package. The extremely low tritium release rates obtained here and the physcial integrity of the solidified product/4/ suggest that a high integrity outer package is not required and that a research program into materials for this package is not necessary.

The accumulation of the basic release data from solidified and encapsulated specimens permits the examination of a number of options in the design of the complete multiple barrier system. The retention of the mold as a barrier to tritium release may eliminate the need for encapsulation of solidified liquid wastes. The solidified liquid wastes and the encapsulated solid wastes, both within their respective molds, may be placed directly into an outer package, such as a standard 200 L drum. The material and shape of the outer package may be dictated more by the type of disposal site available rather than the ability to retain tritium.

TABLE 4

SUMMARY OF TRITIUM RELEASE DATA FROM TRITIATED SPECIMENS IN LIGHT WATER

| Solidification Medium | Encapsulating Material | Specific Dimensions | Surface/ Volume Ratio ($cm^{-1}$) | Tritium Content Original Activity | Leachant Volume (mls) | Leach Time (days) | Average Daily % Tritium Released | Source/ Reference |
|---|---|---|---|---|---|---|---|---|
| Portland Cement Type 10 | None | 5cmDx5cmH* | 1.2 | 2230 μCi | 200 | 180 | $3.5\times10^{-1}$ | Ontario Hydro |
| High Alumina Cement | None | " | " | 2510 | " | 180 | $4.3\times10^{-1}$ | |
| Water Extendible Polyester (Aropol 661-P) | None | " | " | 2685 | " | 180 | $1.3\times10^{-1}$ | Research |
| Portland Cement Type 10 | Epon 815/506 | " | " | 2400 | " | 150 | $1.67\times10^{-3}$ | |
| Portland Cement Type 10 | Epon 815/Lo/Los | " | " | 2305 | " | 150 | $1.40\times10^{-3}$ | Division |
| Portland Cement Type 10 | WEP(Aropol 661-P) | " | " | 2410 | " | 150 | $1.31\times10^{-4}$ | |
| Portland Cement Type 10 | WEP (Aropol 661-P) | " | " | 2180 | " | 60 | $3.38\times10^{-2}$ | (This report) |
| Portland Cement Type 10 | Aropol C-308 Filled | " | " | 2580 | " | 150 | $1.41\times10^{-4}$ | |
| Portland Cement Type 10 | Portland Cement Type 10 | " | " | 2070 | " | 60 | $1.9\times10^{-1}$ | |
| Water Extendible Polyester (Aropol 661-P) | Epon 815/506 | " | " | 2910 | " | 150 | $2.55\times10^{-4}$ | |
| Water Extendible Polyester (Aropol 661-P) | Epon 815/Lo/Los | " | " | 3000 | " | 150 | $2.09\times10^{-4}$ | |
| Water Extendible Polyester (Aropol 661-P) | WEP (Aropol 661-P) | " | " | 2860 | " | 150 | $1.40\times10^{-4}$ | |
| Water Extendible Polyester (Aropol 661-P) | WEP (Aropol 661-P) | " | " | 2805 | " | 60 | $3.72\times10^{-2}$ | |
| Water Extendible Polyester (Aropol 661-P) | Aropol C-308 Filled | " | " | 3210 | " | 150 | $3.35\times10^{-2}$ | |
| Water Extendible Polyester (Aropol 661-P) | Portland Cement Type 10 (10mm layer in each case) | " | " | 2720 | " | 60 | $4.83\times10^{-2}$ | |
| Portland Cement Type 10 | None | 6.67cmDx5.79cmH | 0.94 | 387.0 Ci | 1600 | 122 | $2.5\times10^{-1}$ | Mound ( 5 ) |
| Polymer Impregnated Cement | None | " | " | 386.3 | " | 182 | $1.0\times10^{-1}$ | |
| Portland Cement Type 10 | Polyethylene Bottle | " | NA** | 193.4 | " | 182 | $2.0\times10^{-1}$ | |
| Polymer Impregnated Cement | Polyethylene Bottle | " | NA** | 386.5 | " | 182 | $1.0\times10^{-1}$ | |
| Portland Cement Type 10 | None | 5cmDx5cmH | 1.2 | 320 μCi | 500 | 140 | $3.0\times10^{-1}$ | LASL ( 6 ) |
| Portland Cement Type 10 | 1-2mm Asphalt Coating | " | " | 320 | " | 140 | $5.7\times10^{-2}$ | |
| Portland Cement Type 10 | None | " | " | 1000 | " | 140 | $3.4\times10^{-1}$ | |
| Portland Cement Type 10 | Beeswax or Paraffin Wax Coating | " | " | 1000 | " | 140 | $1.0\times10^{-2}$ | |
| Portland Cement Type 10 | 19-25mm Asphalt Coating | 10cmDx10cmH | 0.6 | ? | 1500 | 28 | $3.1\times10^{-4}$ | |
| 1x Resin/Tritiated Water Mixed with Polyester Resin | None | 5cmDx5cmH | 1.2 | 1200 μCi | ? | 150 | $3.1\times10^{-1}$ | CEA France ( 7 ) |
| 1x Resin/Tritiated Water Mixed with Epoxy Resin | None | " | " | 1200 | ? | 150 | $1.7\times10^{-1}$ | |
| 1x Resin/Tritiated Water Mixed with Polyester Resin | 10mm layer of polyester | " | " | 1200 | ? | 150 | $3.5\times10^{-4}$ | |
| 1x Resin/Tritiated Water Mixed with Epoxy Resin | 10mm layer of epoxy | " | " | 1200 | ? | 150 | $1.0\times10^{-4}$ | |
| Tritiated Water in Hollow Cement Cylinder | 10mm layer of polyester | 8cmDx8cmH | 0.75 | 2515 μCi | ? | 110 | $1.9\times10^{-3}$ | |
| Tritiated Water in Hollow Cement Cylinder | 10mm layer of epoxy | " | " | 2515 | ? | 110 | $1.0\times10^{-5}$ | |

* D - Diameter H - Height

** Specimens were exposed in a polyethylene bottle which was larger than the specimen. Therefore, the surface to volume ratio is not applicable.

TABLE 5

COMPARISON OF TRITIUM RELEASE DATA FROM SPECIMENS EXPOSED TO AIR

| Specification Medium | Encapsulating Material | Specimen Dimensions | Surface/ Volume Ratio ($cm^{-1}$) | Tritium Content µCi | Air Conditions | Leach Time (days) | Average Daily % Tritium Released | Source/ Reference |
|---|---|---|---|---|---|---|---|---|
| Portland Cement Type 10 | None | 5 cm diam x5 cm high | 1.2 | 2050 | flowing | 28 | $2.5 \times 10^{-1}$ | LASL (6) |
| Tritiated Water in Hollow Cement Cylinder | 10 mm layer of epoxy | 8 cm diam x8 cm high | 0.75 | 2515 | flowing | 110 | $4.3 \times 10^{-5}$ | CEA France (7) |
| Tritiated Water in Hollow Cement Cylinder | 10 mm layer of polyester | 8 cm diam x8 cm high | -- | 2515 | flowing | 110 | $2.6 \times 10^{-5}$ | |
| Portland Cement Type 30 | None | 5 cm diam x5 cm high | 1.2 | 2210 | 20°C, 20% rh, 50cc/ min | 60 | $5.9 \times 10^{-1}$ ** | Ontario Hydro |
| Water Extendible Polyester (Aropol 661-P) | None | 5 cm diam x5 cm high | 1.2 | 2635 | " | 60 | $2.1 \times 10^{-1}$ | This report |
| Portland Cement Type 30 | None | 5 cm diam x5 cm high | 1.2 | 1660 | static, 350 cc air space | 120 | $7.0 \times 10^{-5}$ * | Ontario Hydro |
| High Alumina Cement | None | 5 cm diam x5 cm high | 1.2 | 1660 | " | 120 | $8.0 \times 10^{-5}$ * | This report |
| Water Extendible Polyester (Aropol 661-P) | None | 5 cm diam x5 cm high | 1.2 | 1925 | " | 120 | $6.8 \times 10^{-5}$ * | |

* As shown in Appendix 7-4, the tritium levels in the air surrounding the specimens reached a 'constant' value after 1 day. The tests were continued for 120 days but no significant change occurred.

** Specimens split in half.

## CONCLUSIONS

1. Water extendible polyester is superior to Portland cement type 30 and high alumina cement for the solidification and retention of tritiated water.

2. The average daily tritium release rate from solidified but not encapsulated specimens varies from 0.13 to 0.43 per cent per day, is only slightly affected by the choice of solidification medium, and is independent of initial tritium concentration.

3. Exposure of tritiated waste forms prepared with water extendible polyester and Portland cement type 30 to a flowing air stream at 20% relative humidity resulted in tritium release rates similar to those obtained from water immersion.

4. The free space within a sealed container of a solidified tritiated waste will quickly saturate with moisture at a tritium content equal to that of the water within the solidified mass. In the absence of mixing, the tritium content will be reduced proportional to the original relative humidity of the air.

5. Encapsulation of solidified aqueous tritiated wastes within a 10 mm layer of water extendible polyester resin reduces the tritium release rate by a factor of 1000. Epoxy resins were not as effective with the least effective materials being Portland cement and a polyester resin filled with bentonite and sand.

6. Water extendible polyester is the preferred material for solidification and encapsulation of aqueous tritiated wastes and encapsulation of tritiated solids. The use of this material permits a unified approach to conditioning of almost all Ontario Hydro low and intermediate level liquid and solid wastes.

7. The low tritium release rate for solidified and encapsulated wastes would result in the release of <8% of the original activity over 150 years. When allowance is made for the tritium decay only $3 \times 10^{-3}$% of the original activity would be released over 150 years.

8. A high integrity package is not required for solidified and encapsulated tritiated low and intermediate level wastes.

## RECOMMENDATIONS

1. Determine the effect of the specimen surface to volume ratio on tritium release rates by the use of larger specimens up to 20 L in volume.

2. Identify and test suitable mold materials and evaluate the need for an encapsulating layer in addition to the mold.

3. Optimize the thickness of the encapsulating layer with respect to tritium release and minimum volume requirements between the inner and outer packages.

4. Determine tritium release rates from specimens of solidified organic wastes.

5. Determine the effect of higher tritium concentrations on water extendible polyester by solidifying tritiated water containing up to 1000 Ci/L of tritium.

## ACKNOWLEDGEMENT

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Approved:

D. Harrison
Manager
Chemical Research Dept

Submitted:

S.H. Hawthorne
Engineer
Organic Section

SHH:rdm

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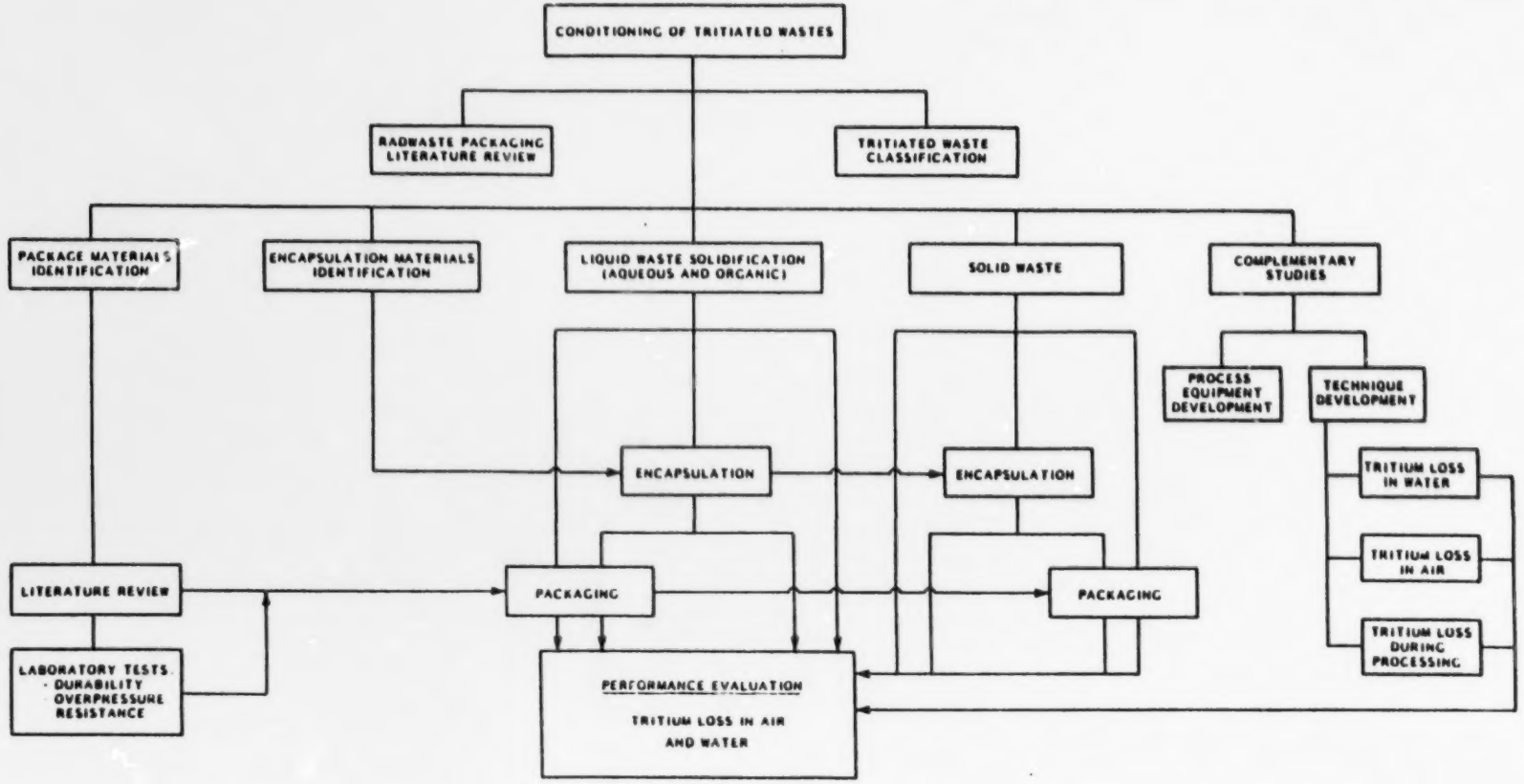
# APPENDIX 1


CONDITIONING OF TRITIATED WASTES
RADWASTE PACKAGING LITERATURE REVIEW
TRITIATED WASTE CLASSIFICATION
PACKAGE MATERIALS IDENTIFICATION
ENCAPSULATION MATERIALS IDENTIFICATION
LIQUID WASTE SOLIDIFICATION (AQUEOUS AND ORGANIC)
SOLID WASTE
COMPLEMENTARY STUDIES
PROCESS EQUIPMENT DEVELOPMENT
TECHNIQUE DEVELOPMENT
ENCAPSULATION
ENCAPSULATION
TRITIUM LOSS IN WATER
TRITIUM LOSS IN AIR
TRITIUM LOSS DURING PROCESSING
LITERATURE REVIEW
PACKAGING
PACKAGING
LABORATORY TESTS
- DURABILITY
- OVERPRESSURE RESISTANCE
PERFORMANCE EVALUATION
TRITIUM LOSS IN AIR AND WATER

## APPENDIX 2

## PREPARATION OF SOLIDIFIED SPECIMENS

### Cement Solidification

#### High Alumina Cement

Cement of known mass was added slowly while stirring into a known mass of tritiated water. A "Jiffy" mixing blade was used at approximately 100 RPM. On obtaining a complete mixture, as evident from the consistency of the cement paste, samples were poured into 120 mL high density polyethylene jars (dimensions 50 mm diameter x 65 mm high) to a height of 50 mm and sealed with screw caps. A batch of material consisting of 1000 g of cement, 300 g of water (0.3/1.0 water/cement ratio) was used to produce 5 specimens at a time in a 1.3 litre container.

An activity of 2200 to 3000 µCi per specimen was obtained by using 50 µCi/mL tritiated water. All specimens and containers were weighed and a theoretical activity was calculated. As a control, one specimen was placed into 1000 mL of demineralized water before it cured and thoroughly agitated. This was allowed to stand 24 h before a 1.0 mL sample was withdrawn for analysis. Results of some of the controls are given in Table 2-1 below.

After 1 or 28 days cure, the specimens were stripped from the molds. All operations were completed in a fume hood.

#### Portland Type 30 Cement

These specimens were prepared using the same method as high alumina cement except a super plasticizer Mighty 150 (sulphonated napthalene formaldehyde) manufactured by KAO, Japan, and distributed by Atkemix, Brantford, was used at a concentration of 20 g/kg of the cement mass.

#### Water Extendible Polyester - Aropol WEP 661-P, Ashland Chemicals

An emulsion consisting of 50:50 by volume of tritiated water and Ashland Aropol WEP 661-P water extendible polyester resin was produced using a high speed mixer and a simple three bladed stirrer at 3000 RPM. The emulsion was formed by pouring the water slowly into the polyester resin while stirring and was then solidified by adding MEK peroxide (90 g/kg active peroxide) at a concentration of 20 g/kg of the polyester resin mass and allowed to cure for 1 day. The specimen container, size and activity was similar to those of the cement sample.

To determine the activity in each specimen, 1 mL of uncatalyzed emulsion was dissolved in 100 mL of methylethylketone and the activity of the solution measured.

TABLE 2-1

| Solidifying Agent | Specimen # | Calculated Activity (mCi) | Measured Activity (mCi) |
|---|---|---|---|
| Type 30 Cement | 35 | 2.21 | 2.18 |
| | E-13 | 2.48 | 2.30 |
| High Alumina Cement | 40 | 2.54 | 2.47 |
| | 45 | 2.47 | 2.46 |
| WEP 661-P | 50 | 2.69 | 2.65 |
| | E-40 | 2.91 | 2.90 |

## APPENDIX 3

## ENCAPSULATION OF SOLIDIFIED SPECIMENS

Tritiated water was solidified using WEP or Type 30 Portland cement to produce a specimen 30 mm in diameter by 30 mm high containing 2200 to 3000 µCi of tritium. After curing for 1 day in sealed polyethylene containers, the specimens were placed into 120 mL polyethylene containers 50 mm in diameter by 65 mm high. The soldified specimens were centered by using a wire basket and the encapsulation materials poured around the smaller specimens to produce a 10 mm thickness of encapsulation material around the entire specimen as detailed in Figure 3-1. After a 7-day cure all specimens were removed from molds and placed into appropriate tests.

The types and manufacturers of the encapsulation materials used are listed below.

| Encapsulation Material | Manufacturer |
|---|---|
| Epon 815 epoxy resin cured with Ancamine 506 amido-amine | Shell Canada Chemical Company |
| Epon 815 epoxy resin cured with a 50:50 mass ratio of Ancamine Lo/Los aromatic amine | Shell Canada Chemical Company |
| Water Extendible Polyester - Aropol WEP 661-P 50/50 water/ WEP by volume | Ashland Chemical |
| Polyester resin Aropol C-308 combined with 1.5 volumes of No 2 sandblasting sand and 0.75 volumes of bentonite clay* | Ashland Chemical |

* Black Hills Bentonite Clay Company

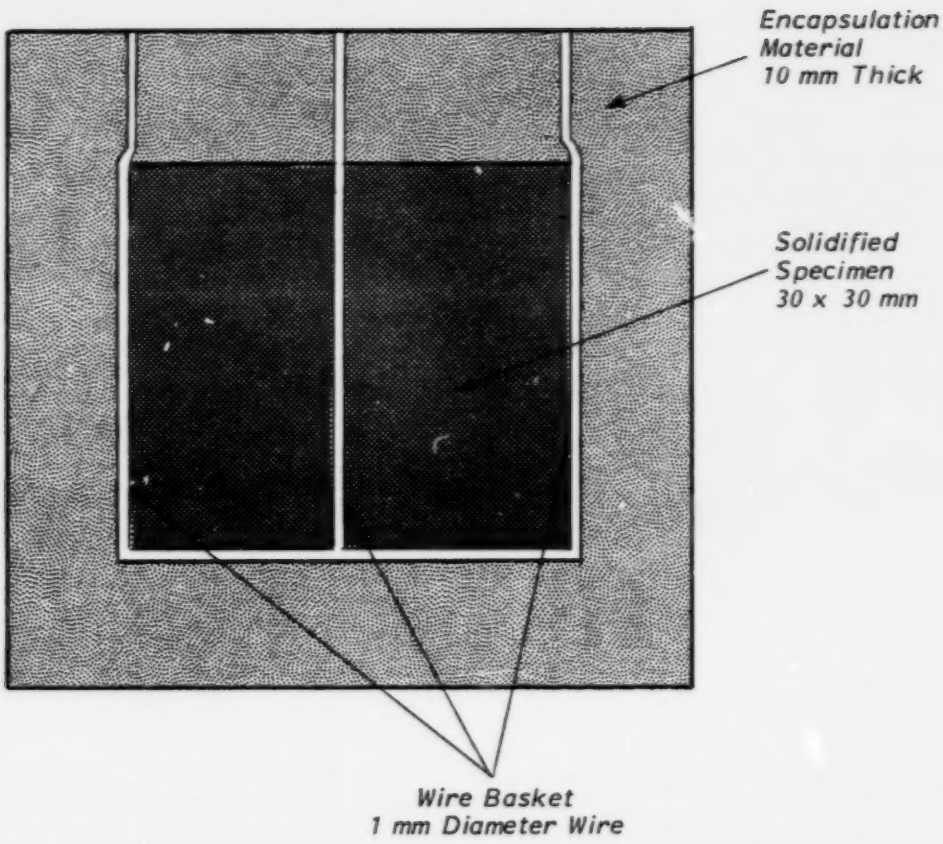



FIGURE 3-1

CROSS SECTION OF ENCAPSULATED
SOLIDIFIED TRITIATED WATER SAMPLE

## APPENDIX 4

## LEACHING IN STATIC WATER

Solidified or solidified and encapsulated specimens were placed into 450 mL glass jars with an internal diameter of 65 mm and a height of 120 mm. Demineralized water (200 mL) was placed into the container to produce a 20 mm head above the specimen. All containers were capped and the leachant was not changed throughout the test.

Samples of the lixivium were taken after 1, 7, 14, 28, 60, 120 and 180 days of leaching and were withdrawn from the solidified specimen containers through rubber septums with a 150 mm long needle and syringe. Caps were removed to obtain samples from the solidified and encapsulated specimen containers. After diluting the 1 mL sample of lixivium to an appropriate activity, 1 mL was placed into 15 mL of PCS scintillation liquid (manufactured by Amersham Corporation) for analysis.

## APPENDIX 5

## LEACHING IN STATIC AIR

Solidified specimens 50 mm in diameter by 50 mm high were placed into a 450 mL glass jar with an internal diameter of 65 mm and a height of 120 mm. All containers were capped and samples were taken through a rubber septum with a 10 mL air-tight syringe and a 35 mm long needle. When obtaining the samples, 1 mL of demineralized water was retained in the syringe to act as a trap when the 9 mL air sample was taken up. After removing the 9 mL of air the syringe was vigorously shaken and the sample was added to 15 mL of PCS (Amersham Corp) scintillation liquid for analysis. An equal quantity of room air was then injected into the sample container to prevent a negative pressure within the specimen container.

## APPENDIX 6

## LEACHING IN FLOWING AIR

Solidified or solidified and encapsulated specimens were placed into a glass jar with inside dimensions, 65 mm diameter by 120 mm high. Specimens were supported on a copper wire stand and an inlet and outlet tube provided on the desorption cell as shown in Figure 6-1. The apparatus was assembled as shown in Figure 6-2 and a flow of 50 cc/min was maintained through the desorption cells. A 20% relative humidity was obtained by regulating the flow of $F_1$ (dry air) and $F_2$ (water saturated air) to obtain an 80/20 ratio respectively. The incoming air, saturation chamber and desorption cells were all maintained at 20°C to prevent condensation inside the air lines. All lines after the desorption cells were metal or glass to limit adsorption of tritium.

The collection bubblers contained 100 mL of ethylene glycol and a 1 mL sample of the collection liquids was taken at various intervals with a 150 mm needle and syringe. In all cases the collection syringe was washed 6 times with clean water between different specimens and all work was completed in a fume hood.

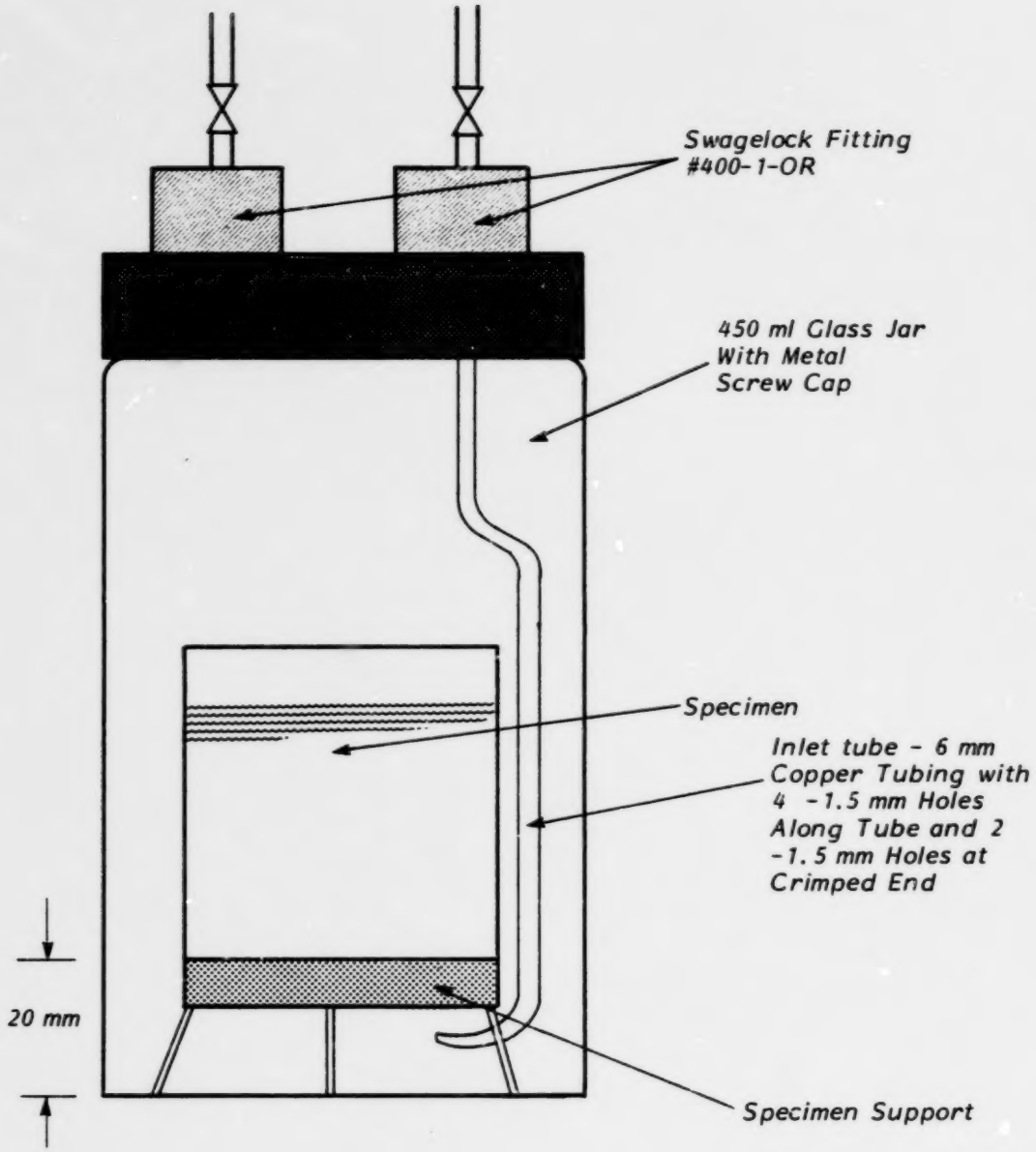



FIGURE 6-1
DESORPTION CELL



84-6
164230 RD

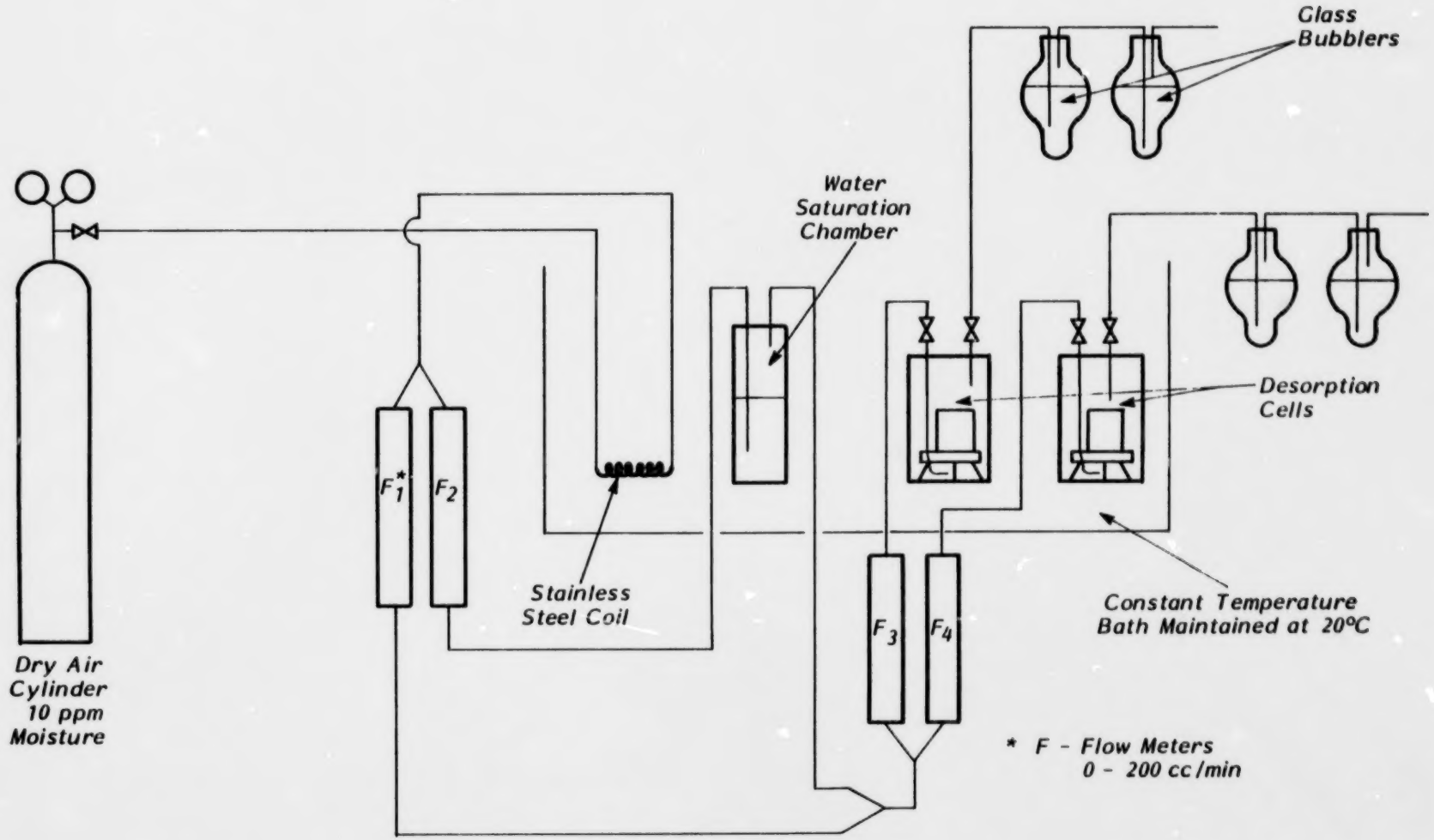



FIGURE 6-2

FLOWING AIR APPARATUS FOR TRITIUM LOSS MEASUREMENTS



84-6
164231 RD

## APPENDIX 7-1

## TRITIUM RELEASES IN WATER FROM SOLIDIFIED TRITIATED AQUEOUS WASTE SPECIMENS

Cure time: 1 day  Tritiated Water Concentration: 51.6 μCi/mL  Temperature: 23°C

| Specimen No | Substrate | Specimen Mass (g) | Water Mass Fraction | Original Activity (μCi) | Cumulative Tritium Releases μCi Time (days) 1 | 2 | 5 | 7 | 14 | 28 | 60 | 120 | 180 |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 26 | Portland Cement | 200.97 | 0.220 | 2280 | 240 | 280 | 400 | 520 | 570 | 830 | 1070 | 1280 | 1430 |
| 27 | Type 30 | 192.02 | 0.220 | 2180 | 240 | 260 | 400 | 490 | 580 | 800 | 1020 | 1250 | 1380 |
| Cumulative Releases as % of original activity | | | | | 10.8 | 12.1 | 17.9 | 22.6 | 25.8 | 36.6 | 46.9 | 56.7 | 63.0 |
| 36 | High Alumina | 214.92 | 0.227 | 2520 | 320 | 520 | 840 | 1040 | 1210 | 1440 | 1800 | 1950 | 1950 |
| 37 | Cement | 213.28 | 0.227 | 2500 | 400 | 580 | 900 | 1100 | 1300 | 1620 | 1850 | 1940 | 1890 |
| Cumulative Releases as % of original activity | | | | | 14.3 | 21.9 | 34.7 | 42.6 | 50.0 | 61.0 | 72.7 | 77.5 | 76.5 |
| 46 | Water Extendible | 98.05 | 0.535 | 2710 | 50 | 60 | 110 | 130 | 170 | 250 | 360 | 520 | 630 |
| 57 | Polyester (WEP) | 96.58 | 0.535 | 2660 | 40 | 60 | 100 | 120 | 160 | 230 | 340 | 480 | 660 |
| Cumulative Releases as % of original activity | | | | | 1.7 | 2.2 | 3.9 | 4.7 | 6.1 | 8.9 | 13.0 | 18.6 | 24.0 |
| Cure time: 28 days  Tritiated Water Concentration: 51.6 μCi/mL | | | | | | | | | | | | | |
| 31 | Portland Cement | 194.08 | 0.219 | 2190 | 200 | 260 | 370 | 430 | 570 | 810 | | 1200 | 1300 |
| 32 | Type 30 | 209.78 | 0.219 | 2370 | 200 | 270 | 380 | 440 | 580 | 810 | | 1200 | 1300 |
| Cumulative Releases as % of original activity | | | | | 8.8 | 11.6 | 16.5 | 19.1 | 25.2 | 35.6 | | 52.7 | 57.1 |
| 41 | High Alumina | 210.62 | 0.232 | 2520 | 450 | 640 | 1020 | 1220 | 1500 | 1770 | | 1800 | 1900 |
| 42 | Cement | 209.78 | 0.232 | 2510 | 440 | 630 | 1010 | 1220 | 1500 | 1810 | | 1800 | 1900 |
| Cumulative Releases as % of original activity | | | | | 17.7 | 25.2 | 40.4 | 48.5 | 59.6 | 71.2 | | 71.6 | 75.5 |

## APPENDIX 7-2

### TRITIUM RELEASES IN WATER FROM ENCAPSULATED SOLIDIFIED TRITIATED AQUEOUS WASTE SPECIMENS

### SOLIDIFIED SPECIMEN DATA

Substrate: Portland Cement Type 30 Tritiated Water Concentration: 214 µCi/mL Temperature: 23°C

Cure time: 1 day

| Specimen No | Encapsulating Material | Solidified[1] Specimen Mass (g) | Water[2] Mass Fraction | Original Activity µCi | Cumulative Tritium Releases µCi Time (days) 7 | 35 | 60 | 120 | 150 |
|---|---|---|---|---|---|---|---|---|---|
| E-1 | Epon 815/506 | 49.8 | 0.219 | 2330 | 1.40 | 3.14 | 3.70 | 5.10 | 5.70 |
| E-2 | | 52.6 | 0.219 | 2470 | 1.74 | 3.54 | 4.14 | 5.58 | 6.30 |
| | Cumulative Releases as % of original activity ($\times 10^2$) | | | | 6.50 | 13.90 | 16.30 | 22.20 | 25.00 |
| E-3 | Epon 815/LO/LOS | 45.7 | 0.219 | 2140 | 0.56 | 1.76 | 2.36 | 3.56 | 4.10 |
| E-4 | | 52.7 | 0.219 | 2470 | 0.70 | 2.28 | 3.14 | 4.60 | 5.62 |
| | Cumulative Releases as % of original activity ($\times 10^2$) | | | | 2.70 | 8.70 | 11.80 | 17.60 | 21.00 |
| E-5 | WEP | 52.3 | 0.219 | 2450 | 0.03 | 0.04 | 0.07 | 0.75 | 1.56 |
| E-8 | (Aropol 661-P) | 50.5 | 0.219 | 2370 | 0.03 | 0.04 | 0.05 | 0.23 | 0.72 |
| | Cumulative Releases as % of original activity ($\times 10^3$) | | | | 1.20 | 1.70 | 2.50 | 20.20 | 47.00 |
| E-9 | Aropol C-308 | 55.7 | 0.219 | 2610 | 0.12 | 0.64 | 2.76 | 26.40 | 59.20 |
| E-10 | filled | 54.5 | 0.219 | 2550 | 0.14 | 0.51 | 1.90 | 21.50 | 50.20 |
| | Cumulative Releases as % of original activity ($\times 10^3$) | | | | 5.00 | 22.30 | 90.10 | 927.30 | 2118.40 |

Substrate: Portland Cement Type 30 Tritiated Water Concentration: 205 µCi/mL

Cure time: 1 day

| | | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| E-24 | Portland Cement | 44.7 | 0.221 | 2030 | 52.0 | 165 | 245 | |
| E-25 | Type 30 | 46.5 | 0.221 | 2110 | 47.0 | 153 | 225 | |
| | Cumulative Releases as % of original activity | | | | 2.4 | 7.7 | 11.4 | |
| E-29[3] | WEP | 46.0 | 0.200 | 2070 | 0.06 | 0.31 | 0.67 | |
| E-30 | (Aropol 661-P) | 50.9 | 0.200 | 2290 | 0.11 | 0.19 | 0.19 | |
| | Cumulative Releases as % of original activity ($\times 10^3$) | | | | 3.90 | 11.6 | 20.3 | |

[1] Mass of solidified but not encapsulated specimens
[2] Mass fraction of water in solidified specimens
[3] Independent replicates of E5 and E8

## APPENDIX 7-3

## TRITIUM RELEASES IN WATER FROM ENCAPSULATED SOLIDIFIED TRITIATED AQUEOUS WASTE SPECIMENS

### SOLIDIFIED SPECIMEN DATA

Substrate: Water Extendible Polyester (Aropol 661-P) Tritiated Water Concentration: 214 µCi/mL

Cure time: 1 day Temperature: 23°C

| Specimen No | Encapsulating Material | Solidified[1] Specimen Mass (g) | Water[2] Mass Fraction | Original Activity µCi | Cumulative Tritium Releases µCi Time (days) 7 | 35 | 60 | 120 | 150 |
|---|---|---|---|---|---|---|---|---|---|
| E-15 | Epon 815/506 | 27.1 | 0.490 | 2840 | 0.22 | 0.53 | 0.66 | 0.99 | 1.03 |
| E-16 | | 28.4 | 0.490 | 2980 | 0.24 | 0.60 | 0.76 | 1.06 | 1.20 |
| Cumulative Releases as % of original activity (x $10^3$) | | | | | 7.9 | 19.4 | 24.4 | 35.2 | 38.3 |
| E-17 | Epon 815/LO/LOS | 28.2 | 0.490 | 2960 | 0.09 | 0.38 | 0.54 | 1.04 | 1.38 |
| E-18 | | 29.0 | 0.490 | 3040 | 0.06 | 0.18 | 0.24 | 0.42 | 0.49 |
| Cumulative Releases as % of original activity (x $10^3$) | | | | | 2.5 | 9.4 | 13.1 | 24.5 | 31.4 |
| E-19 | WEP | 26.9 | 0.490 | 2820 | 0.09 | 0.21 | 0.30 | 0.54 | 0.68 |
| E-20 | (Aropol 661-P) | 27.7 | 0.490 | 2900 | 0.03 | 0.12 | 0.19 | 0.54 | 0.52 |
| Cumulative Releases as % of original activity (x $10^3$) | | | | | 2.1 | 5.8 | 8.6 | 18.9 | 21.0 |
| E-21 | Aropol C-308 | 31.3 | 0.49 | 3280 | 0.08 | 2.10 | 13.2 | 83.4 | 140.58 |
| E-22 | filled | 29.9 | 0.49 | 3140 | 0.03 | 4.54 | 22.0 | 113.0 | 180.76 |
| Cumulative Releases as % of original activity (x $10^2$) | | | | | 0.2 | 10.4 | 55.2 | 307.1 | 502.1 |
| Substrate: Water Extendible Polyester (Aropol 661-P) Tritiated Water Concentration: 205 µCi/mL<br>Cure time: 1 day | | | | | | | | | |
| E-27 | Portland Cement | 26.3 | 0.522 | 2820 | 10.0 | 47.0 | 81.2 | | |
| E-28 | Type 30 | 24.5 | 0.522 | 2620 | 9.4 | 46.0 | 76.3 | | |
| Cumulative Releases as % of original activity | | | | | 0.4 | 1.7 | 2.9 | | |
| E-32[3] | WEP | 24.8 | 0.520 | 2640 | 0.01 | 0.01 | 0.02 | | |
| E-33 | (Aropol 661-P) | 27.9 | 0.520 | 2970 | 0.01 | 0.06 | 0.11 | | |
| Cumulative Releases as % of original activity (x $10^4$) | | | | | 3.6 | 12.0 | 22.3 | | |

[1] Mass of solidified but not encapsulated specimens
[2] Mass fraction of water in solidified specimens
[3] Independent replicates of E19 and E20

## APPENDIX 7-4

### TRITIUM RELEASES IN STATIC AIR FROM SOLIDIFIED TRITIATED AQUEOUS WASTE SPECIMENS

Cure time: 1 day  Tritiated Water Concentration: 35.4 µCi/mL  Temperature: 23°C

| Specimen No | Substrate | Specimen Mass | Water Mass Fraction | Original Activity µCi | Cumulative Tritium Releases µCi Time (days) | | | | |
|---|---|---|---|---|---|---|---|---|---|
| | | | | | 1 | 5 | 14 | 28 | 120 |
| 54 | Portland Cement Type 30 | 211.00 | 0.217 | 1620 | 0.14 | 0.14 | 0.14 | 0.14 | 0.14 |
| 55 | | 221.05 | 0.217 | 1700 | 0.14 | 0.14 | 0.15 | 0.13 | 0.14 |
| Cumulative Releases as % of original activity ($\times 10^3$) | | | | | 8.4 | 8.4 | 8.7 | 8.1 | 8.4 |
| 57 | High Alumina Cement | 204.30 | 0.229 | 1660 | 0.16 | 0.16 | 0.17 | 0.17 | 0.16 |
| 58 | | 204.50 | 0.229 | 1660 | 0.18 | 0.15 | 0.15 | 0.17 | 0.16 |
| Cumulative Releases as % of original activity ($\times 10^3$) | | | | | 10.2 | 9.3 | 9.6 | 10.2 | 9.6 |
| 60 | Water Extendible Polyester | 101.60 | 0.533 | 1920 | 0.12 | 0.14 | 0.15 | 0.14 | 0.15 |
| 61 | | 102.23 | 0.533 | 1930 | 0.13 | 0.13 | 0.14 | 0.14 | 0.16 |
| Cumulative Releases as % of original activity ($\times 10^3$) | | | | | 6.5 | 7.0 | 7.5 | 7.3 | 8.1 |
| Cure time: 28 days  Tritiated Water Concentration: 51.6 µCi/mL | | | | | | | | | |
| 33 | Portland Cement Type 30 | 202.98 | 0.219 | 2290 | 0.22 | 0.16 | 0.22 | 0.22 | |
| 34 | | 207.20 | 0.219 | 2340 | 0.20 | 0.18 | 0.20 | 0.21 | |
| Cumulative Releases as % of original activity ($\times 10^3$) | | | | | 9.1 | 7.4 | 9.1 | 9.3 | |
| 43 | High Alumina Cement | 205.00 | 0.232 | 2450 | 0.23 | 0.20 | - | 0.21 | |
| 44 | | 205.80 | 0.232 | 2460 | 0.19 | 0.17 | 0.18 | 0.21 | |
| Cumulative Releases as % of original activity ($\times 10^3$) | | | | | 8.6 | 7.5 | 7.3 | 8.6 | |

## APPENDIX 7-5

### TRITIUM RELEASES IN FLOWING AIR FROM SOLIDIFIED TRITIATED AQUEOUS WASTE SPECIMENS

Air Conditions: 20°C, 20% relative humidity, 50 mL/min  Tritiated Water Concentration: 50.7 µCi/mL

Cure time: 1 day

| Specimen No | Substrate | Specimen Mass | Water Mass Fraction | Original Activity µCi | Cumulative Tritium Releases µCi Time (days) | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | | | 1 | 2 | 5 | 14 | 28 | 60 | 75 |
| 69 | Portland Cement Type 30 | 185.18 | 0.229 | 2150 | 32.7 | 65.0 | 150 | 321 | 472 | 722 | 850 |
| 70 | | 195.25 | 0.229 | 2270 | 32.2 | 60.4 | 146 | 327 | 531 | 849 | 1018 |
| Cumulative Releases as % of original activity | | | | | 1.5 | 2.8 | 6.7 | 14.7 | 22.7 | 35.5 | |
| 72 | Water Extendible Polyester | 99.25 | 0.520 | 2620 | 27.0 | 44.0 | 85.0 | 149 | 214 | 342 | 404 |
| 73 | | 100.41 | 0.520 | 2650 | 25.8 | 42.0 | 85.0 | 147 | 217 | 312 | 372 |
| Cumulative Releases as % of original activity | | | | | 1.0 | 1.6 | 3.2 | 5.6 | 8.2 | 12.4 | |